Path Distinguishability in Double Scattering of Light by Atoms

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Wave-particle duality finds a natural application for electrons or light propagating in disordered media where coherent corrections to transport are given by two-wave interference. For scatterers with internal degrees of freedom, these corrections are observed to be much smaller than would be expected for structureless scatterers. By examining the basic example of the scattering of one photon by two spin-$\frac{1}{2}$ atoms—a case-study for coherent backscattering—we demonstrate that the loss of interference strength is associated with which-path information stored by the scattering atoms.

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Einstein's and de Broglie's wave-particle duality (WPD)—the ability of a quantum system to display the seemingly contradictory attributes that one would have regarded as wave-like and particle-like and, therefore, as mutually exclusive in pre-quantum physics—is arguably the most important phenomenological consequence of Bohr's principle of complementarity. The quantitative aspects of WPD are particularly well understood [1] in the context of two-paths interferometers where a definite path is particle-like and the interference between the amplitudes of the two paths is wave-like.

The wave-like interference strength is quantified by the familiar Michelson's fringe visibility $\mathcal{V}$, and the particle-like path knowledge is measured by the path distinguishability $\mathcal{D}$, which is perhaps less familiar and has this operational meaning: The odds for guessing the path right are $(1+\mathcal{D})/2$. The extreme situations of no path knowledge and high fringe visibility (ideally $\mathcal{D}=0$, $\mathcal{V}=1$) or full path knowledge and no fringes ($\mathcal{D}=1$, $\mathcal{V}=0$) are standard textbook fare. A rather recent experiment with a bearing on the matter discussed below is the one carried out by Eichmann *et al.* in 1993 [2].

The compromises allowed by the laws of physics in intermediary situations are restricted by the duality relation [3, 4]

$$\mathcal{D}^2+\mathcal{V}^2\leq 1\,. \tag{1}$$

It is worth noting that well pronounced wave-like and particle-like aspects can coexist: With odds for guessing the path right of 99% ($\mathcal{D}=0.98$), we can have well visible fringes of 20% visibility ($\mathcal{V}=0.2$).

The two pioneering experiments that tested the duality relation employed two-path interferometers for atoms [5] and photons [6], whereby internal degrees of freedom of the interfering objects themselves were used to provide the path information. By contrast, in the situation that we examine here—coherent double scattering—the which-path information is stored in the deflecting elements of the interferometer (atoms) and not carried by the interfering objects (photons).

We wish to show how these notions of path knowledge and interference strength are naturally applied to coherent wave transport. In the semi-classical regime of weak localization, most of the coherent effects in wave transport can be explained by *two-wave* interference between amplitudes propagating in opposite direction along loop-like scattering paths [7, 8]. These interference corrections to transport can dramatically alter the diffusion process and even suppress it [9, 10]. They are sensitive to several "dephasing" processes but most of them are circumvented at sufficiently low temperatures [11, 12, 13].

Here we address an intrinsic dephasing mechanism which survives at zero temperature: the path knowledge stored in the internal degrees of freedom of the scatterers. Indeed, when the scatterers have an internal structure, the interference corrections to transport are observed to be rather small, for example in the scattering of electrons by magnetic impurities at very low temperatures [12].

The same effect has been observed in the coherent backscattering (CBS) of light by cold rubidium atoms [14, 15, 16]. This coherent multiple scattering effect arises when an optically thick sample of scatterers is illuminated by coherent light. It, too, results from interference of light amplitudes, here of the two amplitudes associated with traversing the same path in opposite direction. The endpoints of each scattering path thus play the role of Young slits and give rise to an angular fringe pattern in the far-field. Owing to the varying separation between the endpoints, these patterns have different fringe spacings but they *all* display a bright fringe at backscattering. Thus, the sum of all fringe patterns displays an angular peak around the backscattering direction [17].

We quantify the strength of this interference in a natural manner by the relative excess of the peak intensity over the background, the analog of the fringe visibility $\mathcal{V}$ in this context. For atoms with a spin-0 ground state, this CBS peak-to-background ratio reaches its maximal possible value of 2 in the helicity-preserving polarization channel [18], corresponding to $\mathcal{V}=1$, whereas it is very

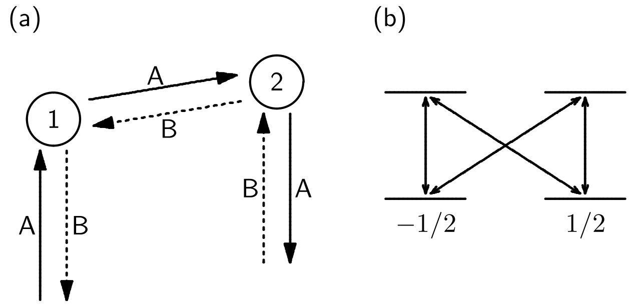

FIG. 1: (a) The two paths in coherent backscattering. Along path A the photon is first scattered by atom 1, then by atom 2; along path B the order is reversed. (b) Level scheme of the $\frac{1}{2} \leftrightarrow \frac{1}{2}$ transition. Both the ground state and the excited state are doublets with total angular momentum $j = \frac{1}{2}$, and the magnetic quantum numbers $m = \pm\frac{1}{2}$ label the sublevels.

small for atoms with a degenerate ground state [14], as is the situation with a Zeeman fine structure or a hyperfine structure.

Our point is that these results can be recast and understood, both qualitatively and quantitatively, in terms of wave-particle duality. Indeed, when scattering the photon the atoms may undergo a change in their ground state—a circumstance equally crucial in the single-scattering situation of the Eichmann *et al.* experiment [2]. This is to say that the atoms can store which-path information so that the experimenter can find out, in principle if not in practice, which of the two atoms scattered first and which second. As a consequence, the strength of the coherent corrections to transport is bounded by the distinguishability of the paths inside the sample, and the height of the CBS peak is limited by the amount of path knowledge available.

We consider the simplest possible scenario that exhibits the effect: double scattering off two identical spin-$\frac{1}{2}$ atoms (atom 1 and atom 2), with the photon resonant with a $\frac{1}{2} \leftrightarrow \frac{1}{2}$ dipole transition. In fact, this is the situation of the Eichmann *et al.* experiment where the scatterers are Hg^+ ions. This geometry is simplest for multiple scattering to set in and is at the heart of the CBS phenomenon. Since our focus is on the influence of the internal atomic structure, we assume that the atoms are so stiffly trapped that there is no relevant contribution from the atomic recoil (the storage of CBS which-path information in the center-of-mass degrees of freedom of mobile atoms is studied in Ref. [19]). Put differently, we take for granted that the atomic center-of-mass degrees of freedom do not store which-path information.

To simplify the problem further, we assume that the distance between the atoms is sufficiently large for the double scattering contribution to dominate over all other multiple scattering processes (triple, quadruple, ...). As illustrated in Fig. 1, path A is the case when atom 1 scatters first and atom 2 second (sequence $1 \to 2$); path B is the sequence $2 \to 1$. The paths are geometrically identical but traversed in opposite directions. The two atoms together compose the path detector: the change of their internal states bears witness of the actual path.

Since the ground states are spin-$\frac{1}{2}$ states, the path detector is a qubit pair, which is a 4-state system. During the scattering process, however, the excited states of the atoms are involved as well, and the details of the scattering interaction determine the over-all effect on the atoms. This net before-to-after change in the combined ground states of both atoms is given by a completely positive two-qubit map.

We establish this map by first recalling that the atom-photon interaction is described by quasi-resonant point-dipole elastic scattering. The corresponding transition operator is proportional to $\mathcal{T} = (\boldsymbol{d}\,\boldsymbol{d}) \otimes |\boldsymbol{r}\rangle\langle\boldsymbol{r}|$ where $\boldsymbol{r}$ is the atom's position vector and $\boldsymbol{d}$ is the dipole vector operator of the atomic transition. The omitted proportionality factor depends on the oscillator strength of the transition. It determines the probability of the double scattering event and is, therefore, crucial for an actual experiment. But in the present context this probability is irrelevant because *the final two-atom state is conditioned on successful double scattering.* Bearing this conditioning in mind, we consistently leave all further proportionality factors implicit.

For an incoming photon with wave vector $\boldsymbol{k}$ and transverse polarization $\boldsymbol{\epsilon}$, the matrix elements of $\mathcal{T}$ are

$$\langle m', \boldsymbol{k}'\boldsymbol{\epsilon}'|\mathcal{T}|m, \boldsymbol{k}\boldsymbol{\epsilon}\rangle = \langle m', \boldsymbol{\epsilon}'|(\boldsymbol{d}\,\boldsymbol{d})|m, \boldsymbol{\epsilon}\rangle\; e^{i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{r}}\,, \quad (2)$$

where $\boldsymbol{k}'$ and $\boldsymbol{\epsilon}'$ are the wave vector and polarization of the outgoing photon, and m and m' are the magnetic quantum numbers of the initial and final ground state, respectively [16]. Since the scattering is elastic, we have $|\boldsymbol{k}| = |\boldsymbol{k}'|$.

The dyadic operator $T = (\boldsymbol{d}\,\boldsymbol{d})$ acts on the internal degrees of freedom of the photon (the polarization states) and of the atom (the magnetic sublevels of the angular momentum multiplets). Its matrix elements read

$$\langle m', \boldsymbol{\epsilon}'|(\boldsymbol{d}\,\boldsymbol{d})|m, \boldsymbol{\epsilon}\rangle = \langle m'|(\boldsymbol{\epsilon}'^{*} \cdot \boldsymbol{d})(\boldsymbol{d} \cdot \boldsymbol{\epsilon})|m\rangle\,. \quad (3)$$

The matrix elements of the vector operator $\boldsymbol{d}$ are the Clebsch–Gordan coefficients that characterize the coupling of spin-1 (photon) with spin-$\frac{1}{2}$ (ground state) to give spin-$\frac{1}{2}$ (excited state); all the coefficients have equal magnitude for such a $\frac{1}{2} \leftrightarrow \frac{1}{2}$ transition. As a consequence, we have effectively $T = (\boldsymbol{\sigma}\,\boldsymbol{\sigma})$ for initial and final ground states, where $\boldsymbol{\sigma}$ is the Pauli vector operator for the spin-$\frac{1}{2}$ ground state. [20]

We consider the exact backscattering geometry where $\boldsymbol{k} = -\boldsymbol{k}'$, which we choose parallel to the z axis of the coordinate system; the magnetic quantum numbers $\pm 1/2$ in Fig. 1(b) also refer to the z direction. For path A, the double scattering operator that acts on the two atomic

ground-state qubits is

$$\begin{aligned} T_{\mathrm{A}} &= \boldsymbol{\epsilon}'^{*}\cdot(\boldsymbol{\sigma}_2\boldsymbol{\sigma}_2)\cdot(\mathbf{1}-\boldsymbol{n}\boldsymbol{n})\cdot(\boldsymbol{\sigma}_1\boldsymbol{\sigma}_1)\cdot\boldsymbol{\epsilon} \\ &= -\boldsymbol{\epsilon}'^{*}\cdot\boldsymbol{\sigma}_2\,(\boldsymbol{\sigma}_2\times\boldsymbol{n})\cdot(\boldsymbol{n}\times\boldsymbol{\sigma}_1)\,\boldsymbol{\sigma}_1\cdot\boldsymbol{\epsilon}\,, \end{aligned} \tag{4}$$

where $\mathbf{1}-\boldsymbol{n}\boldsymbol{n}$ is the dyadic projector onto the plane orthogonal to the unit vector $\boldsymbol{n}$ that points from one scatterer to the other. The double-scattering operator T_{B} for path B is obtained by interchanging $1\leftrightarrow 2$ in (4).

With ρ_{in} denoting the initial two-qubit state of the two scattering atoms, the final states are then given by

$$\begin{aligned} \rho_{\mathrm{A,B}} &= \frac{T_{\mathrm{A,B}}\,\rho_{\mathrm{in}}\,T_{\mathrm{A,B}}{}^{\dagger}}{w_{\mathrm{A,B}}} \\ \text{with}\quad w_{\mathrm{A,B}} &= \mathrm{tr}\bigl\{T_{\mathrm{A,B}}\,\rho_{\mathrm{in}}\,T_{\mathrm{A,B}}{}^{\dagger}\bigr\}\,, \end{aligned} \tag{5}$$

where the normalizing denominators take care of all the proportionality factors that we left implicit. The weights of the two paths are $w_{\mathrm{A}}/(w_{\mathrm{A}}+w_{\mathrm{B}})$ and $w_{\mathrm{B}}/(w_{\mathrm{A}}+w_{\mathrm{B}})$, respectively. In addition to the initial two-atom state ρ_{in}, these weights and the final states depend on the preselected polarization $\boldsymbol{\epsilon}$ of the incoming photon and the post-selected polarization $\boldsymbol{\epsilon}'$ of the outgoing photon, on which the ensemble of events is conditioned.

Since the final states of the atoms are different for the two paths, there is which-path information stored in the atoms, which—in principle—can be extracted by a suitable measurement, although in practice it could be very difficult to implement such a measurement. The optimal measurement would provide as much path knowledge as is available, quantified by the distinguishability of the paths, which is given by [1, 3, 4]

$$\begin{aligned} \mathcal{D} &= \frac{\mathrm{tr}\bigl\{\bigl|w_{\mathrm{A}}\rho_{\mathrm{A}}-w_{\mathrm{B}}\rho_{\mathrm{B}}\bigr|\bigr\}}{w_{\mathrm{A}}+w_{\mathrm{B}}} \\ &= \frac{\mathrm{tr}\bigl\{\bigl|T_{\mathrm{A}}\,\rho_{\mathrm{in}}\,T_{\mathrm{A}}{}^{\dagger}-T_{\mathrm{B}}\,\rho_{\mathrm{in}}\,T_{\mathrm{B}}{}^{\dagger}\bigr|\bigr\}}{w_{\mathrm{A}}+w_{\mathrm{B}}}\,. \end{aligned} \tag{6}$$

This is supplemented by the visibility

$$\mathcal{V} = \frac{2\Bigl|\mathrm{tr}\bigl\{T_{\mathrm{A}}\,\rho_{\mathrm{in}}\,T_{\mathrm{B}}{}^{\dagger}\bigr\}\Bigr|}{w_{\mathrm{A}}+w_{\mathrm{B}}}\,, \tag{7}$$

the quantitative measure for the interference strength of the two paths. Irrespective of the detailed form of ρ_{in} and the operators $T_{\mathrm{A}}, T_{\mathrm{B}}$, the duality relation (1) is obeyed by this distinguishability and visibility [1].

We now restrict the discussion to symmetric initial two-qubit states of the form $\rho_{\mathrm{in}} = \frac{1}{4}(\mathbb{1}-p\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2)$ with $-\frac{1}{3}\le p\le 1$ as required by the positivity of ρ_{in}. This one-parameter family of initial states encompasses some cases of particular physical interest: the completely mixed state ($p=0$); the projector on the singlet state of vanishing total angular momentum ($p=1$); the projector on the triplet sector of unit total angular momentum ($p=-\frac{1}{3}$). It is worth recalling that two-qubit states of this form are separable for $p\le\frac{1}{3}$ and entangled for $p>\frac{1}{3}$ but, as illustrated by Eqs. (9) below, nothing remarkable happens to $\mathcal{D}$ and $\mathcal{V}$ at the transition from $p<\frac{1}{3}$ to $p>\frac{1}{3}$.

For all values of p, the initial state ρ_{in} is invariant under the interchange $1\leftrightarrow 2$ and, therefore, the interferometer is symmetric in the sense that both paths occur with equal *a priori* probability ($w_{\mathrm{A}}=w_{\mathrm{B}}$). As a consequence, the difference of operators in (6) is *antisymmetric* under $1\leftrightarrow 2$ and thus of the form

$$\frac{T_{\mathrm{A}}\,\rho_{\mathrm{in}}\,T_{\mathrm{A}}{}^{\dagger}-T_{\mathrm{B}}\,\rho_{\mathrm{in}}\,T_{\mathrm{B}}{}^{\dagger}}{w_{\mathrm{A}}+w_{\mathrm{B}}} = \boldsymbol{a}\cdot(\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)+\boldsymbol{b}\cdot(\boldsymbol{\sigma}_1\times\boldsymbol{\sigma}_2) \tag{8}$$

with two numerical vectors $\boldsymbol{a}$ and $\boldsymbol{b}$ that depend on the photon polarizations $\boldsymbol{\epsilon},\boldsymbol{\epsilon}'$, the unit vector $\boldsymbol{n}$, and the initial-state parameter p. The right-hand side in (8) is a rank-2 operator with its nonzero eigenvalues given by $\pm2\sqrt{\boldsymbol{a}^2+\boldsymbol{b}^2}$, and so we get $\mathcal{D}=4\sqrt{\boldsymbol{a}^2+\boldsymbol{b}^2}$ for the distinguishability of the paths.

In this manner we arrive at explicit expressions for $\mathcal{D}$ and $\mathcal{V}$ [21, 22]. We will report the full technical details elsewhere and focus here on the particular situation in which the line connecting the two atoms in Fig. 1(a) is perpendicular to the incoming and outgoing propagation directions, that is: we choose the unit vector $\boldsymbol{n}$ along the x axis. For this perpendicular geometry, one has

$$\begin{aligned} \mathcal{D} &= \frac{1+p+2pu}{2(1+pu)}\sqrt{1-u'^2}\,, \\ \mathcal{V} &= \frac{\bigl|(1+p)(1+uu')-2p(1-u')\bigr|}{2(1+pu)}\,, \end{aligned} \tag{9}$$

where u and u' are the x components of the Stokes vectors associated with the incoming and outgoing photon polarizations [23]. These are such that

$$\mathcal{D}\le\begin{cases}\sqrt{(1-\mathcal{V})\mathcal{V}} & \text{for } p\le 0\,,\\ \sqrt{(1-\mathcal{V})\bigl(2p/(1+p)+\mathcal{V}\bigr)} & \text{for } p\ge 0\,.\end{cases} \tag{10}$$

Clearly, the duality relation (1) is obeyed for all p values. The relation is only saturated for $p=1$, in which case the initial atomic state is pure and the equal sign is expected to hold in (1) on general grounds [4].

For the completely-mixed initial state ($p=0$) we have

$$\mathcal{D}=\frac{1}{2}\sqrt{1-u'^2}\,,\qquad \mathcal{V}=\frac{1}{2}(1+uu')\,. \tag{11}$$

Here, the distinguishability does not depend at all on the initial polarization—a surprising feature that is particular to the $\frac{1}{2}\leftrightarrow\frac{1}{2}$ transition in the perpendicular geometry and is not generic. This observation about the perpendicular geometry can be understood as follows.

Since all Clebsch–Gordan coefficients are of equal size, the first scatterer has uniform *a priori* probability of reaching either one of its ground states, irrespective of

the polarization of the incoming photon. Yet, when conditioned on the direction into which the photon is reemitted, the probability is not uniform as a rule, but it *is* for the perpendicular geometry. Therefore, there is no which-path information stored in the final completely-mixed state of the atom that scatters first.

When the observed polarization of the outgoing photon is an equal-weight superposition of the in-plane and out-of-plane linear polarizations ($u' = 0$), the final state of the second scatterer is a corresponding pure state. So, when finding only one atom in this pure state, we can infer the path with certainty, but if both atoms are found in this state, we know nothing about the path and will guess wrong half of the time. Both cases are equally probable, so that our betting odds are 75%, which is consistent with $\mathcal{D} = \frac{1}{2}$ for $u' = 0$ in (11), as it should be. In this case the visibility is $\mathcal{V} = \frac{1}{2}$ irrespective of the incoming polarization.

The distinguishability is zero for an outgoing photon linearly polarized in the plane of the drawing in Fig. 1(a), when $u' = 1$, or perpendicular to this plane ($u' = -1$). The corresponding visibility takes on any value between 0 and 1. The case $\mathcal{V} = 0$ happens for photons with perpendicular polarizations, one with in-plane polarization, the other perpendicular ($uu' = -1$). The case $\mathcal{V} = 1$ occurs for photons with parallel linear polarizations, both in the plane or both perpendicular to it ($uu' = 1$).

Let us now turn to the situation of an initial singlet state ($p = 1$). As noted above, the duality relation (1) is then saturated and we have

$$\mathcal{D} = \sqrt{1 - u'^2}\,, \qquad \mathcal{V} = \left|u'\right|. \tag{12}$$

The fact that the distinguishability does not depend on the initial polarization can be understood by an argument similar to the one given above for the $p = 0$ case. Irrespective of the incoming photon polarization and for, say, a left-circular outgoing photon, the final states of the atoms have $(m_1, m_2) = (\frac{1}{2}, -\frac{1}{2})$ for path A and $(-\frac{1}{2}, \frac{1}{2})$ for path B whereas the reversed situation occurs for a right-circular outgoing photon. This means that if the outgoing light is analyzed in the circular channels ($u' = 0$), perfect path knowledge is available ($\mathcal{D} = 1$) and no interference will be observed ($\mathcal{V} = 0$). Conversely, if the outgoing light is analyzed in the channels of linear in-plane and out-of-plane polarization ($\left|u'\right| = 1$), no path knowledge is available ($\mathcal{D} = 0$) and one recovers full interference strength ($\mathcal{V} = 1$).

Finally, one can think of mimicking the physics of the CBS phenomenon by an angular average over the direction $\boldsymbol{n}$. We first calculate the average of the difference operator in (8) and then compute the resulting distinguishability as the trace of its modulus. The corresponding visibility is obtained as the angular average of the $\boldsymbol{n}$ dependent visibility (7). For $p = 0$, which applies to most of the available experimental data, the largest averaged distinguishability is $\mathcal{D} = \frac{1}{2}$; it is found in the helicity-preserving polarization channel. The smallest average visibility is also found in this channel, its value is $\mathcal{V} = \frac{2}{5}$. Even if a direct quantitative comparison with the real CBS situation cannot be made at this stage, this result is nevertheless consistent with the experimental observation that the lowest CBS peaks are actually found in this detection channel [14].

In summary, we have demonstrated that the concept of wave-particle duality proves relevant and useful for our understanding of some aspects of the interference effects in multiple scattering. To make solid quantitative contact with actual CBS experiments, the analysis must be extended to account for scattering by three and more atoms. The stage for this future research is now set.

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